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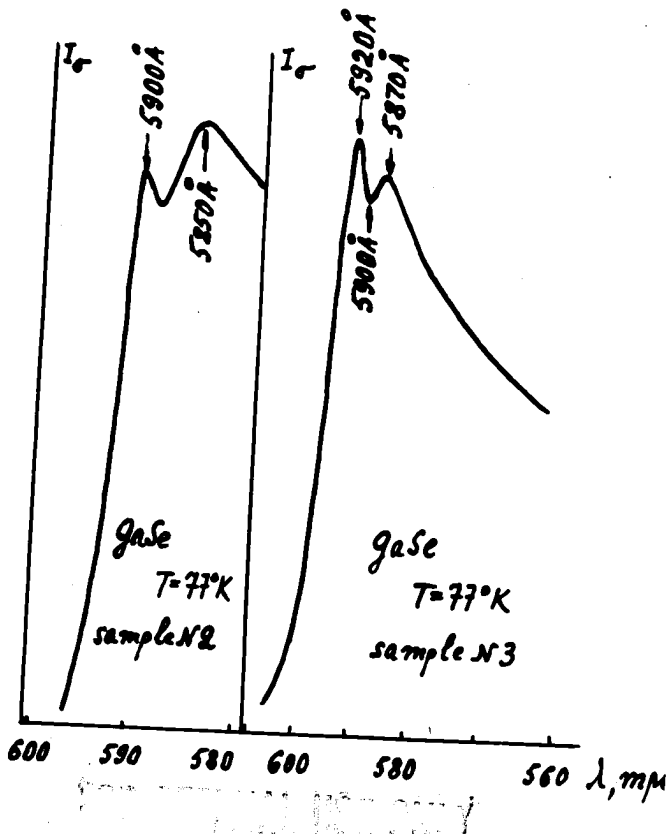

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Fig 5

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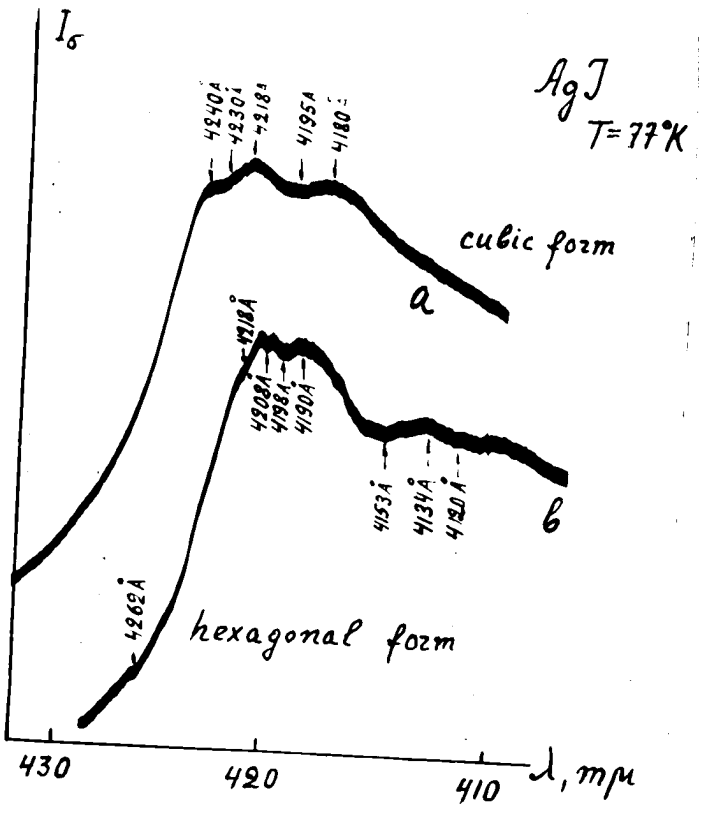

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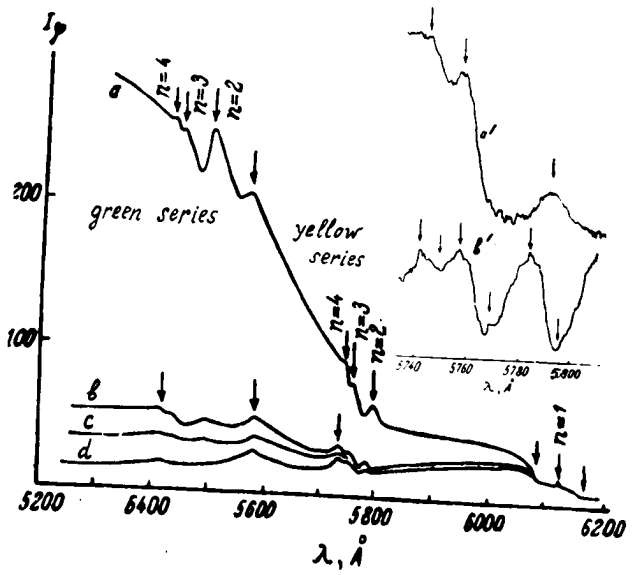

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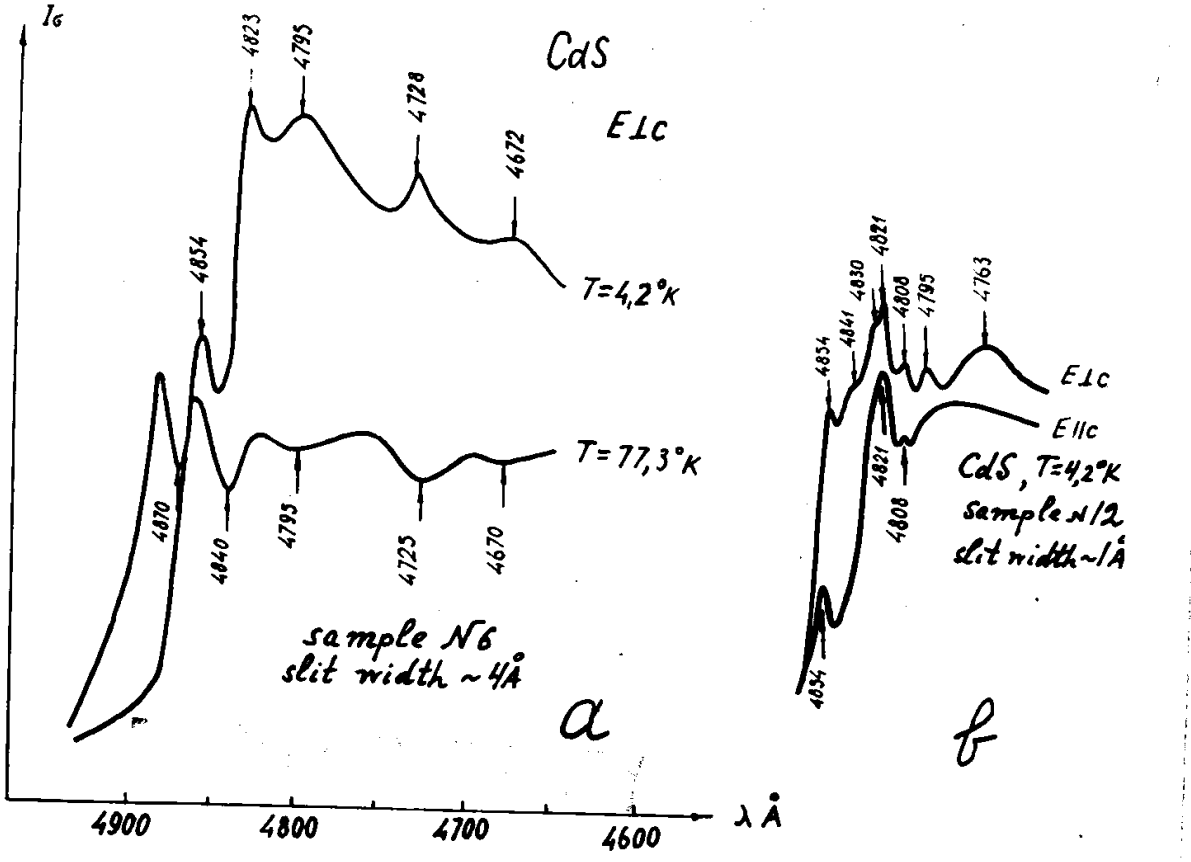

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Fig 12

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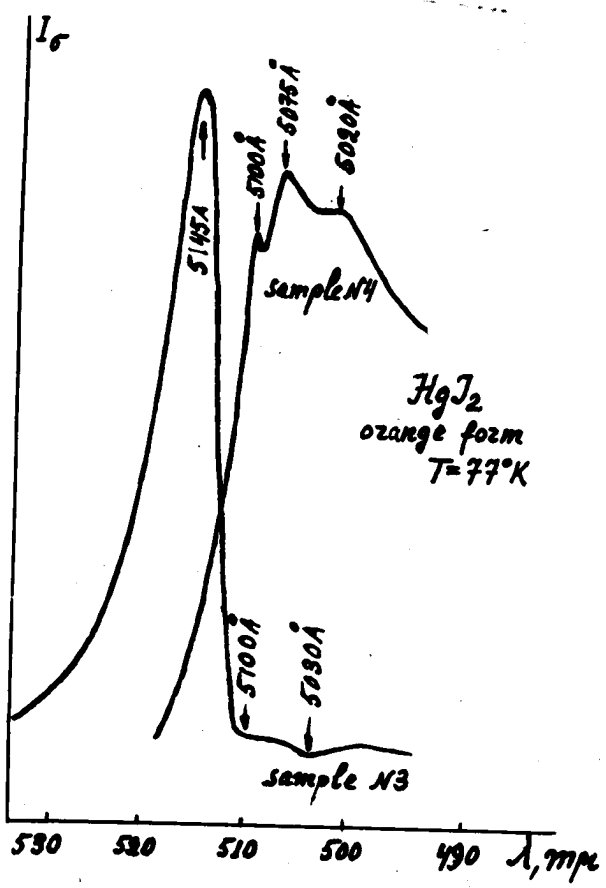

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Fig 6

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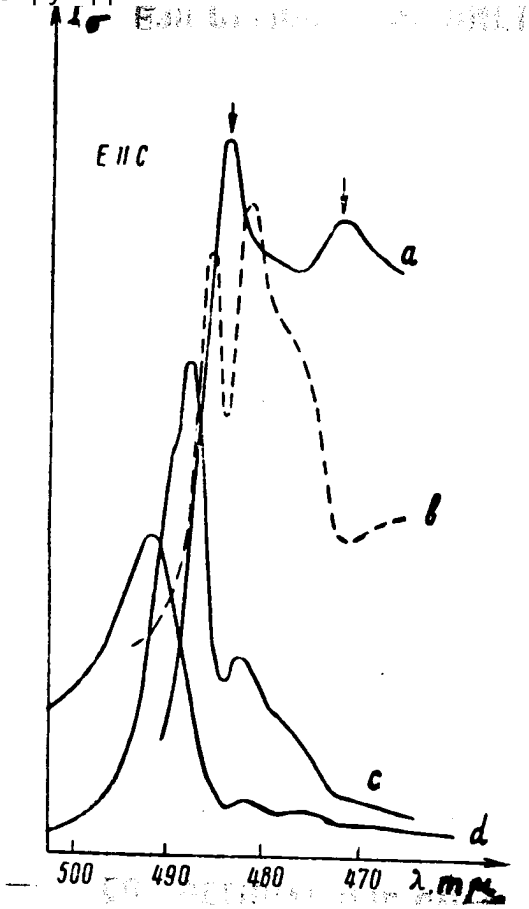

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Fig 10

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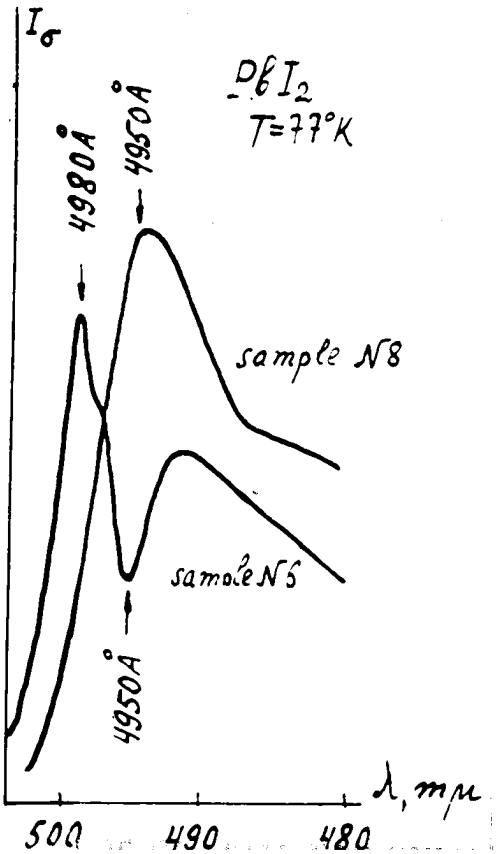

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Fig V

Gross E.F and Novikov B.V

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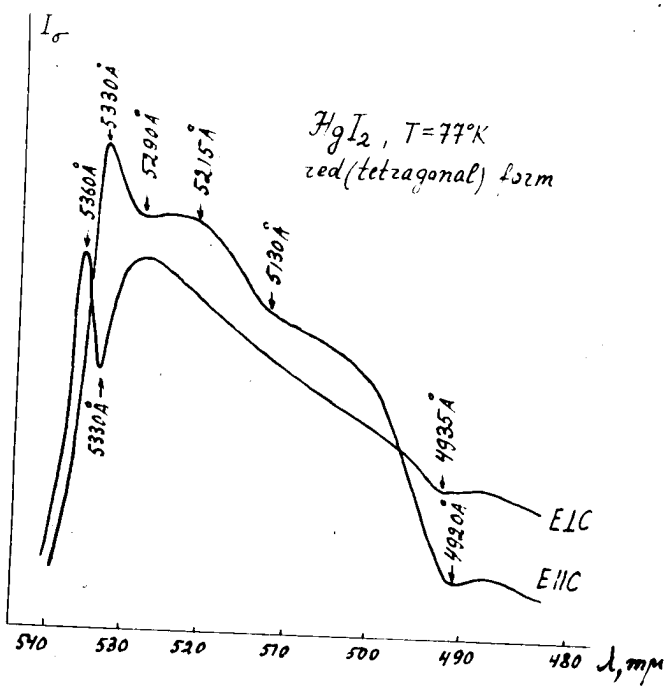

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Fig 5
Gross S.F and Norinder BN

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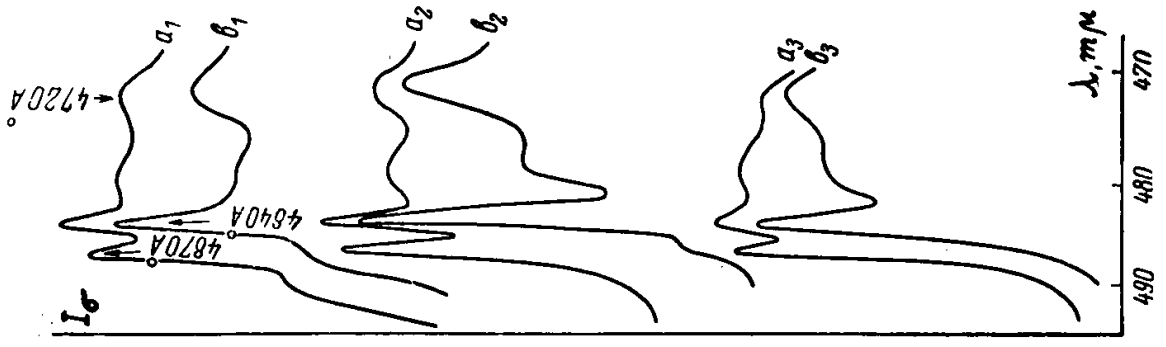

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Gross E. F and Nevikov B.V

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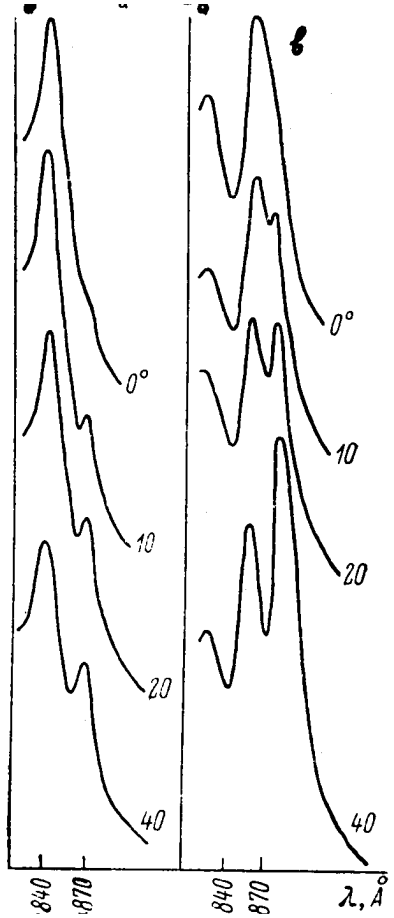

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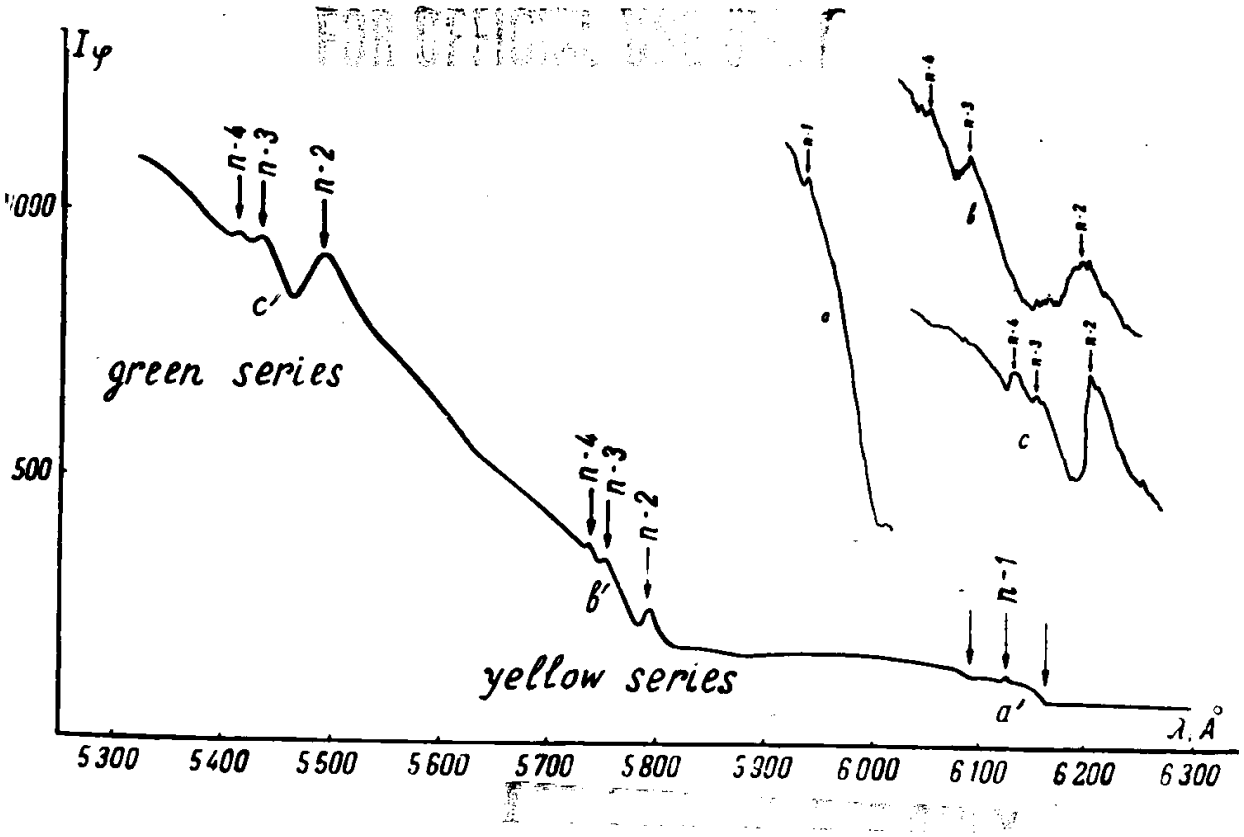

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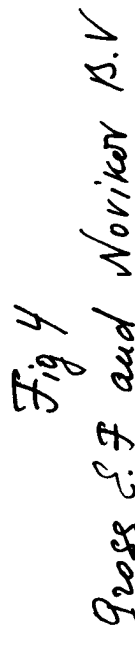

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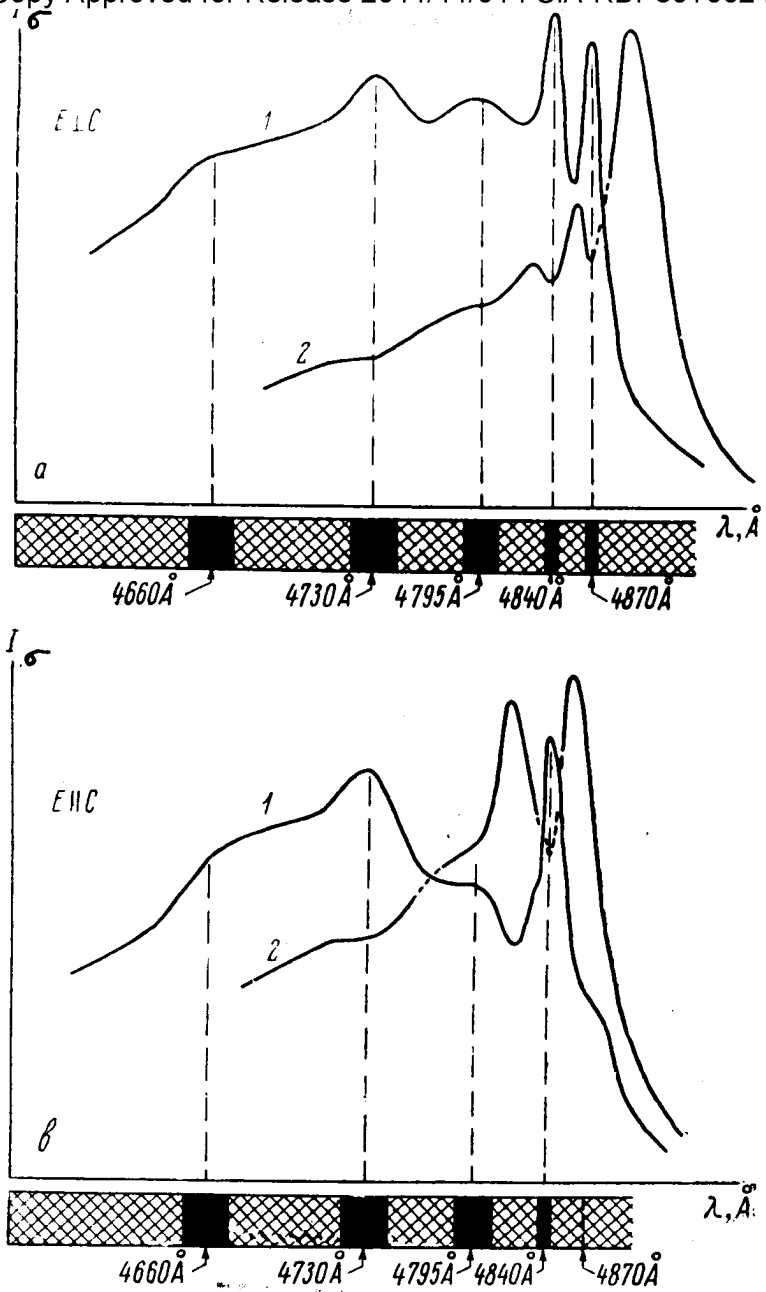

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Fig 1

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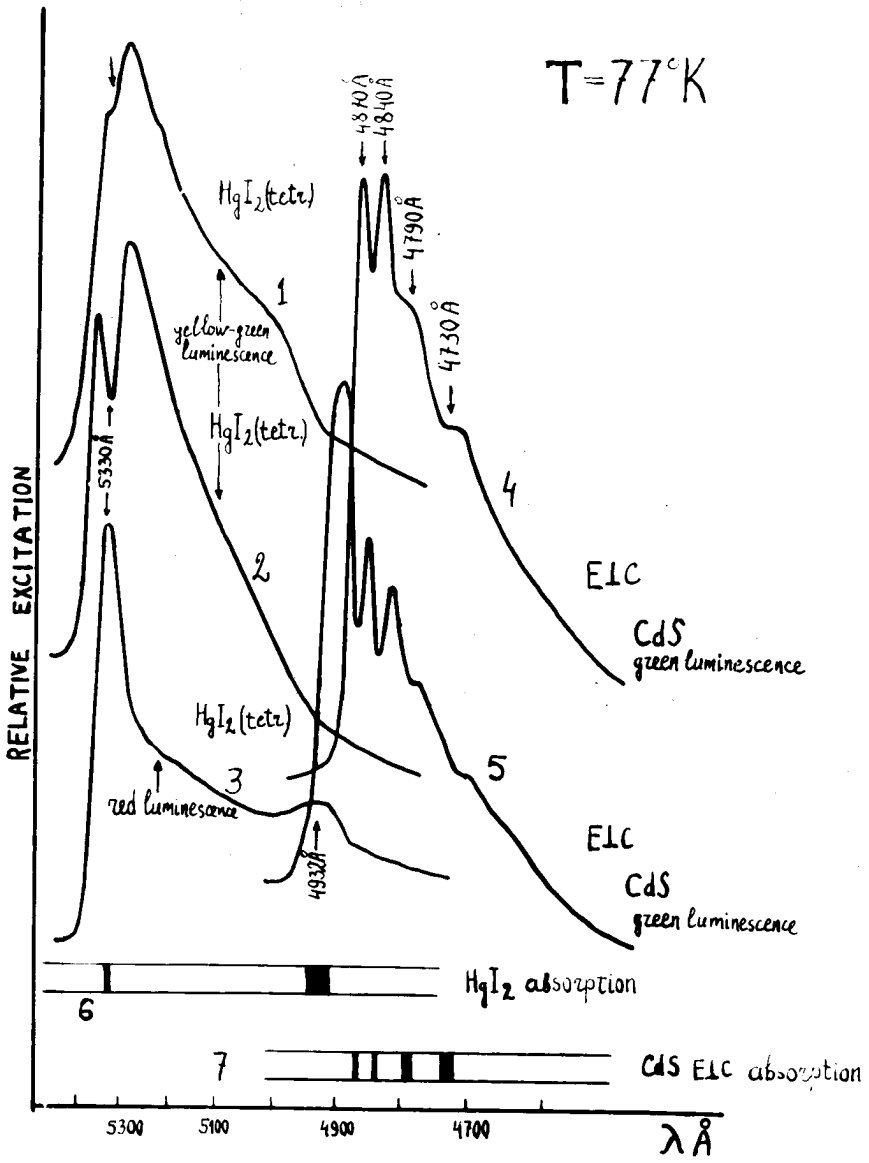

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Fig 13

Gross E.F and Noviker B.V.

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THE FINE STRUCTURE OF THE SPECTRAL CURVES OF PHOTOCONDUCTIVITY AND LUMINESCENCE EXCITATION AND ITS CORRELATION TO EXCITON ABSORPTION SPECTRUM

E. F. Gross and B. V. Novikov

ABSTRACT

The exciton fine structure of the fundamental absorption edge in semiconductor crystals was examined. It was observed that substances having such a structure are fairly strong photoconductors. This fact leads to the conclusion that there is a correlation between the exciton absorption lines and the shape of photoconductivity curve.

Crystals with well known structures of the absorption edge - CdS, Cu_2O, HgJ_2, PbJ_2 and GaSe were chosen for investigation. A fine structure of the spectral curves of photoconductivity, corresponding to exciton absorption lines, was discovered in all the investigated crystals at low temperatures. Its properties in a CdS single crystal were studied in detail. It turned out that either the maxima (1st type of crystals) or the minima (2nd type of crystals) on the photocurrent curves could correspond to the exciton absorption lines. By means of proper treatment crystals can be converted from one type into another and vice versa.

The coincidence of photocurrent and absorption maxima is considered as a consequence of exciton participation in giving rise to photocurrent. Possible mechanisms of exciton participation in creating charge carriers are discussed.

On the spectral curve of luminescence excitation of CdS crystals at low temperatures, a fine structure related to the exciton spectrum is also observed. Either maxima or minima on the excitation spectral curve coincide with the exciton absorption lines.

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E. F. Gross and B. V. Novikov

In 1931 J. F. Frenkel /1/, for the first time, suggested a hypothesis concerning nonphotoconductive excitation in a crystal lattice. This excitation, capable of migrating through the crystal, was called an exciton.

Frenkel's ideas found further development in the theoretical works of Wannier /2/, Mott /3/, Heller and Marcus /4/, Pekar /5,6/ and others. Later, both theoreticians and experimentators sometimes applied to the hypothesis on the exciton in connection with the attempt to explain some or other phenomena. Thus, for instance, the well-known wide absorption peaks in alkali-halide crystals were identified with excitation, by means of light, of excitons in these crystals. The concept of the exciton was also referred to in connection with migration of energy in luminescense processes /7/. In essence, the concept of the exciton (under the name of "energy migration") was used by Vavilov for the explanation of some phenomena of luminescence in liquids. The role of the exciton as the energy carrier was taken into account when explaining a number of experimental facts in the phenomena of external and internal photoeffect. It was shown in a number of very interesting and important researches of prof. Apker and his collaborators /8/ that a whole series of peculiarities in the phenomenon of photoemission becomes clear, if one assumes the participation of the exciton in these phenomena.

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As a result of a whole series of experiments, carried out for the purpose of studying photoconductivity in Cu_2O crystals, Zhuze and Rivkin /9/ came to the conclusion that the role of the exciton in the phenomenon of photoconductivity is essential. However, the problem whether such a quasi-particle really existed in crystals remained to be proved. Confirmation to the actual existence of such a particle in crystals was obtained further from another quarter - that of optical phenomena, Proceeding from the experimental results of Zhuze and Rivkin, research on optical absorption at low temperatures in Cu_2O crystals was undertaken by one of the authors (E.G.) in 1950, These investigations resulted in the discovery of a hydrogen-like series of narrow lines on the absorption edge of the crystal /10/. This series of lines was interpreted /10/ as the optical exciton spectrum in the Cu_2O crystal.

This investigation, proceeding from the exciton concept, gave an interpretation of the coefficients of the hydrogen-like series /10/, and revealed their relation to the reduced effective mass of exciton (the effective masses of electron and hole), Ridberg's constant and crystal refraction index, on the one hand, and the energy of exciton photo-dissociation - on the other. Further studies [42] as well as detailed electro-optical [35] and magneto-optical [36] investigations of the Cu_2O absorption spectrum confirmed the correctness of its exciton interpretation.

The existence of the hydrogen-like series on a Cu_2O absorption edge was independently discovered also in the

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experiments of Hayashi /11/.[x/] In further investigations, carried out in different laboratories, the complex structure of the absorption edge was observed in a number of other crystals. This phenomenon, which reveals itself with particular distinctness in Cu_2O, is a general property of crystal lattices. At present, there are no doubts that this structure is the exciton optical spectrum of the crystals.

As will be seen further, the complex structure on the absorption edge turned out to be closely connected with the photo-electric phenomena in crystals. First of all, it was noticed /13/ that all crystals with a complex absorption edge structure are strong photo-conductors, e,g. Cu_2O, CdS, CdSe, ZnS, HgI_2, PbJ_2, AgI and others. All these materials at low temperatures reveal narrow and strong lines at the absorption edge; these lines should be connected with the formation of excitons. Besides, the fact that these groups of exciton lines are disposed in that part of the spectrum where the maximum photoconductivity is usually observed, draws attention to itself.

Side by side with crystals in which a clear exciton spectrum

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x/ In this paper /11/, however, Hayashi does not give any interpretation of the hydrogen-like series coefficients, does not relate the hydrogen-like Cu_2O spectrum with the exciton. After the publication of our paper on the exciton spectrum /10/ Hayashi and Katsuki, in a later article[12]subscribed to our interpretation of the hydrogen-like Cu_2O spectrum only in the case of the green series. The Cu_2O yellow series is attributed by them to the polaron, and in a recent 1959 publication /34/ Hayashi again stresses this conception.

One cannot agree with the Japanese authors' latter interpretation of the Cu_2O yellow series as a polaron spectrum, because such an explanation is at variance with experimental facts.

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with strong lines is observed, there are crystals which do not reveal the absorption edge structure, e.g.: MoO_3, Bi_2O_3, V_2O_5, As_2S_3, PbO (the yellow form) and others. It was noticed that in such crystals photoconductivity is either not observed at all or is of a very low value.

The correlation between the exciton lines and photoeffect displays itself particularly clearly in the case of red and yellow modifications of HgI_2 crystals /14/. In the red modification there are exciton lines and a strong photoeffect; in the yellow modification the exciton lines are absent and the photoeffect is extremely weak. This fact is particularly convincing, as the correlation between the exciton lines and the photoconduction value is observed here on one and the same material.

A detailed study of the spectral photosensitivity of crystals, in the absorption spectra of which lines, connected with exciton, are observed, is of great importance for clearing up the role of excitons in photoconductivity. The discovery of photosensitivity peaks, coinciding with exciton absorption lines, would be direct proof that formation of excitons at light absorption leads to the appearance of charge carriers.

The first experiments in this direction were undertaken by us together with A.A.Kaplianski in 1955 on CdS and HgI_2 crystals /15/. Both these materials are strong photoconductors and have a rich structure in absorption. At present, the absorption edge spectrum of CdS /31-33/ has been studied in particular details. The theoretical investigations of Birman /16/ and Hopfield /17/ made it possible to classify the inten-

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sive exciton lines into three groups - A, B and C, each of which corresponds to the transition to exciton levels from one of the valence bands.

Investigations on the spectral distribution of photocurrents in monocrystals CdS were carried out at T=77°K and 4°K with polarized light /18/. A mono-chromator of a relatively high dispersion was used for the investigations. The width of the slit varied within the limits from 0.5 A to 5 A. The thickness of the CdS samples (and all other investigated by us substances) was chosen with the aim in mind so that all the amount of light in the spectral region of the exciton lines was fully absorbed by the crystal.

About 80 monocrystal samples, grown in different ways, were investigated.[x/] At low temperatures the spectral photocurrent curves of all the investigated crystals reveal clear peaks in the spectral range of exciton absorption lines. The shape of the photocurrent curves and its character of correlation with the absorption lines, make it possible to divide the large variety of obtained curves into two types.

In certain samples (conventionally attributed to the "first" type) the photocurrent maxima (peaks) correspond to the absorption lines on the spectral distribution curve and coincide in position with them.

In other samples (the "second" type) the spectral photocurrent curve differs greatly from the curve for crystals

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x/ The authors are grateful to Prof. S.M.Rivkin (Physico-Technical Institute, Leningrad) and Prof. R.Rompe (Institut fur Strahlungsquellen Berlin) for the possibility of experimentation with crystals grown in their laboratories.

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of the first type: in this case, the absorption lines coincide in position with the photocurrent minima (dips). The spectral photocurrent curves, for crystals of both types, depend considerably upon the orientation of the crystal with respect to the direction of propagation and polarization of the incident light.

In fig. 1 characteristic spectral photocurrent curves obtained on two samples belonging to different types are shown, for example. The curves were obtained for different polarizations of incident light.

In the long-wave part of the curve of first-type crystals (fig 1, a and b, curves 1) there is a number of clearly defined peaks. The first of them 4870 A is polarized with the electric vector $E \perp C$. This peak, according to position and polarization, coincides with the most long-wave intensive absorption line in the crystals CdS observed at $T = 77^{\circ}K$. The remaining photocurrent peaks (some of which, apparently, are only partially polarized) are located near $\lambda\lambda$ 4840 A, 4795, 4730 and 4660 A . - These peaks also correspond to the intensive absorption lines the position of which is shown in fig 1. Both long-wave photocurrent peaks 4870 A and 4840 A are rather narrow, their half-width is $6 \div 8$ A .

In fig. 1 it is seen that the photocurrent curve of 1st type crystals looks like a wide, smooth maximum ("background") with protruding peaks above it. The relative value of peaks and background, on which these peaks are located, may greatly differ in various samples. In some samples the peaks make a comparatively small addition to the strong background

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(curves a_3b_3 in fig 2), while as in others it is the other way round: the largest part of sensitivity is concentrated in the peaks (curves a_2b_2 in fig 2). It is noteworthy that an increase of the background leads to a decrease of the relative value of peaks, i.e. the background is not simply added to the peaks, but, in a way, absorbs them.[20]

In 2nd type crystals (fig. 1 a and b, curves 2) the steep rise of the photocurrent for both polarizations begins at noticeably larger wavelengths in comparison with the already described 1st type crystals. An essential difference in the crystals of both types is observed also in the short-wave part of the curves. The 2nd type crystals have a steeper short-wave drop of the photo-current curve, where a distinct structure consisting of narrow peaks and dips is observed. The narrow minima (with halfwidth $6 \div 8$A) correspond to absorption lines $\lambda\lambda$ 4870A and 4840A , the wider ones are located near absorption lines $\lambda\lambda$ 4795 and 4730A. In certain samples the minima also is seen near line 4660A . It is of essential importance that in second type crystals the minima (dips) and not the maxima (peaks) on the photocurrent curve correspond to the absorption lines. This is also a significant difference between the two types of crystals.

As is the case in the 1st type crystals, the relative depth and clarity of the photocurrent minima may differ from sample to sample, and, sometimes, may differ also in different parts of one and the same crystal.

Investigations of 2nd type crystals with polarized light reveal a strong dependence of the long-wave part of the photo-

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conductivity curve upon the polarization of the incident light.

The dip on the photo-current curve (fig.1 a, curve 2), connected with line 4870A , is present only in component E⊥C, and is absent in component E // C; this is due to the strong polarization of this absorption line with E⊥ C. It must be noted that strong photocurrent peaks near 4890A on curve 2 (fig 1, a) for component E⊥ C do not correspond to any absorption line. The same takes place also in the case of the most longwave and intensive peak near 4860A for component E // C (fig 1 b, curve 2). Other peaks on the photo-current curves of 2nd type crystals (fig.1, a and b, curve 2) do not have any correspondence with absorption lines either. These peaks, the position of which, by the way, considerably changes from sample to sample, are closely connected with the depth of the minima (dip) on the photo-current curves of 2nd type crystals. These peaks apparently are caused by the side parts ("wings") of the absorption line shape, the central parts of which coincide with the dips of the photo-current curve. Thus, the situation here is analogous to the phenomenon of selfreversion of the atomic spectral lines, when on the shape of the emission line there sometimes also appear two side peaks, which change their position in dependence with the conditions of the experiment. In this respect, the above-mentioned peaks on the photo-current curve of a 2nd type crystal must be regarded as some "false" peaks, which should not correspond to the absorption lines, and, therefore, no definite physical sense should be ascribed to them.

Such a self-reversion of the peak on the spectral curve

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of the photoemission of coloured alkali-halide crystals was observed under certain conditions by Apker and Taft /8/.

At room temperature the difference between 1st and 2nd type crystals still remains. Further we shall consider the connection between these groups, and the conditions under which crystals pass over from one type to the other.

Our following observations are testimony to the fact that the absorption lines are correctly compared with the narrow photo-current peaks (in cases of 1st type crystals) and narrow dips (in 2nd type crystals) /18/.

By orientating the optical axis of the crystal perpendicular to the rotation axis, and parallel to the electric vector of the polarizer, it is possible as is known /32/ to observe, in an extraordinary ray, an intensification of absorption line 4870A when increasing angle ($\hat{E C}$). between the crystal axis and electric vector of the incident light /32/.

As it is seen in fig 3, an increase of the angle between E and C from 0° to 40° in 1st type crystals leads to the appearance of a peak on the photo-current curve in the same place as absorption line 4870A .

When increasing angle ($\hat{E C}$) - the peak increases and the whole curve approaches the shape characteristic of E C.

In fig. 3,b the behaviour of the long-wave part of the photo-current spectral curve for 2nd type crystals at an analogous rotation of the crystal is presented. In these crystals, when increasing the angle between E and C on the long-wave drop of the photo-current curve (on the drop of the "false" peak), a dip, which rapidly increases with the increase of the angle, appears in the same place as absorption line

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4870 A. As a result of this there appears on the photo-current curve a second "false" peak, which shifts in the direction of the larger wave lengths, and grows in size while rotating the crystal.

At large angles the shape of the curve approaches the curve characteristic of E⊥C in 2nd type crystals also.

It is known that when lowering the temperature to 4°K, the absorption edge of CdS crystals, and the lines located near it shift in the short-wave direction. The spectral photo-current curves also shift in the same direction. The narrow long-wave photo-current peaks in 1st type crystals at T = 4°K become yet narrower and take up a position near λλ 4853 and 4823 AA ,(i.e. are displaced from their positions at T = 77° by 17A) fig. 12b. It is possible to observe also at 4°K a photo-current peak at 4808A. In 2nd type crystals the photo-current dips are located at the same wave-lengths. The wide, short-wave photo-current peaks (dips) undergo a somewhat smaller temperature shift.

The insufficient dispersion of the monochromator did not enable us so far to reveal the photo-current peaks' fine structu re of which is connected with the existence of longitudinal and transverse excitons /21/ in CdS crystals. According to the theoretical conception of Pekar and Dykman /22/ the longi-tudinal exciton waves must give rise to a photo-current several times stronger than in the case of transverse ones.

A cooling to 4°K considerably influences the general shape with the spectral photo-current curve (see further).

A question naturally arises whether the discovered pheno-menon (the fine structure of the photoconductivity curve) is

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a general one for all crystals with an exciton spectrum. The most interesting substance from this point of view was Cu_2O, where the exciton absorption spectrum is observed especially clearly. Indeed, the experiments carried out in the Soviet Union (Gross - Pasternjak /23/) and in the United States (Apfel-Portis /24/) proved that on the Cu_2O photo-current curve, in the yellow and green parts of the spectrum, photo-current peaks are observed. They form two hydrogen-like series, which exactly correspond to the yellow and green series of the exciton in Cu_2O (see fig. 4). At the same time, the shape of the photo-conductivity curve reflects all the peculiarities of the absorption edge structure.

A similar conformity between the fine absorption structures and photo-conductivity has been discovered now in a large number of substances. Nearly simultaneously with CdS these phenomena were discovered by us in a crystal of red modification - HgI_2. Investigations were carried out on a number of single HgI_2 crystals in polarized light. The most typical curves are presented in fig. 5. A comparison of the absorption spectra data /14,26/ reveals that on the photo-current curve in the case of $E \perp C$ the dips at $\lambda\lambda$ 5330A and 4930A coincide with the absorption lines. According to our terminology this curve belongs to the 2nd type of crystals. Apparently, the dips on photo-current curve for the case of extraordinary ray ($E \parallel C$) should also be compared with the absorption lines. Absorption line 5330A is polarized with $E \perp C$, and, therefore, the structure only on the photo-current curve for ordinary ray should correspond to it. The narrow photo-current peak in component E C, situated near line 5330A, we think, should not be

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connected with the absorption line. This peak (its position differs from sample to sample) is, apparently, a false peak, typical of 2nd type crystals, as is the case in CdS. It must be noted that in some crystals in component $E \perp C$, the photo-current peak may correspond to absorption line 5330A (1st type of crystals).

Side by side with the red modification, the orange modification of HgI_2 also has a strong photo-conductivity. At T = 77°K in the long-wave part of the spectral photo-current curves of these crystals a structure is also observed, the extremes of which may be compared with the absorption lines discovered in this material by Sieskind and his collaborators /25/. In fig.6 two most typical spectral photo-current curves for two different HgI_2 crystals of orange modification are presented.

In single crystals PbI_2 and Ga Se the structure of the absorption edge is particularly simple. Thus, in PbI_2 at T = 77°K a wide band, with 4950A as the centre /14,27/, is observed; in GaSe at the same temperature we may observe a strong line near 4900A, and a wide weak band at 5870-5800 A /28/. The simplicity of the absorption structure in these substances makes it possible to observe, with great obviousness and convincingness, the phenomenon which was for the first time discovered by us with CdS: the absorption lines manifest themselves as photo-current peaks in 1st type crystals, and as dips on the spectral photo-current curves in 2nd type crystals. This phenomenon for PbI_2 and GaSe substances is illustrated in fig. 7 and 8.

We have discovered a rich structure of photo-current curves in polycrystalline films of AgI. Fig 9 a and b gives the photo-

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current curves for the cubic and hexagonal modifications of this substance. A comparison of obtained results with published data on absorption /29/ shows that the dips on the photo-current curves coincide with the absorption lines in both AgI modifications.

The discovered correlation, in a large number of substances, between the structure of the spectral photo-conductivity curves and the line structure of the absorption edge, we think, shows a possibility of using photo-conductivity as a spectral method for investigating energy levels in a crystal. The investigation of energy absorption levels by means of the photo-conductivity method, does away with the difficulties connected with the size, surface conditions, and the thickness of the samples. It is well known that the preparation of very thin single crystals, for investigating their absorption spectrum, presents considerable difficulties. The necessity of a registering apparatus falls away. This fact is very important when investigating those parts of the spectrum, which are difficult for access. As our investigations show, photo-conductivity spectra in some cases are more "sensitive" than the absorption spectra. Thus, in CdS a number of photo-current peaks was discovered considerably earlier than the corresponding lines in absorption.

Thus, the described observations show that the absorption lines are closely connected with the structure of the spectral photo-current curves, can promote both an increase of the photo-current (in the case of peaks), and its decrease (in the case of dips). It presented certain interest to find out under what conditions the absorption lines manifest themselves

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in the spectral distribution of photo-current as peaks, and under what conditions as dips. One of the experiments carried out in this direction was the thermal treatment of CdS crystals with different cooling conditions (annealing and tempering).

CdS crystals were heated in a furnace at T = 400-500°C during 1 - 1 1/2 hours, and then quickly cooled to room temperature, or, more often, to the temperature of liquid nitrogen (tempering). In cases of annealing the crystals, heated during the same period of time, were slowly cooled (approximately 10-12 hours) to room temperature. The spectral photo-current curves were investigated both before and after the described thermal treatment at T = 77°K.

First-type crystals, where the absorption lines correspond to the photo-current peaks, essentially changed the general shape of the spectral photo-current distribution after being subjected to tempering. From the long-wave side there appeared a wide, clear peak; from the short-wave side a redistribution of photo-current values took place in such a way that now the dips on the photo-current curve corresponded to the absorption lines. Such a transition took place with greater certitude at quicker cooling to T = 77.3°K. Tempering did not produce any considerable effect upon 2nd type crystals.

Further we established that 1st type crystals after annealing retained the shape of their spectral curves which they had before the thermal treatment (after annealing in some cases the peaks on the photo-current curve became clearer). In 2nd type crystals after annealing the spectral photo-current curves changed, and took the shape characteristic of 1st-type crystals. Thus, annealing transferred 2nd-type crystals into 1st type

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crystals.

The process of transferring CdS crystals from one type into another is quite reversible. We managed to transfer a 1st type crystal after tempering into a 2nd type crystal, and then by means of annealing to transfer it back to its original state.

Besides annealing and tempering, we investigated the influence of a mechanical treatment of the surface of CdS crystals upon the structure of their photo-current curves /19/. It was possible to presume that the influence of defects, located on the surface, would be particularly essential. The absorption coefficient in the exciton lines and background is quite considerable (more than $10^5 cm^{-1}$), and, consequently, the excitons and free current carriers arise in the immediate proximity of the surface.

In fig. 10 are shown the spectral photo-current curves of a crystal which originally belonged to the 1st type (fig.10a) after different mechanical treatment of the surface. The treatment of the sample surfaces was carried out in the following sequence: polishing on the surface of wet silk (fig. 10 b); polishing on silk with chromium oxide (fig. 10c); grinding with fine emery powder (fig. 10d). An examination of the obtained results makes it possible to mention the following regularities:

1) Already after a slight treatment of the surface the photo-current peaks (curve a) self-reverses, forming dips (curve b, fig.10).

2) The long-wave false peak widens and shifts in the direction of the long waves (curves b,c,d, fig.10) when increasing the degree of surface treatment.

It must be noted that with the increase of degree of surface

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treatment the general photo-sensitivity of the samples also considerably falls.[x/]

In the case of Cu_2O crystals a mechanical treatment of a surface (and also etching of the surface, absorption of water and spirit molecules on it) produces a strong influence on the shape of structure of photo-current curves in the region of Cu_2O exciton series /30/. The photo-conductivity peaks shift, and the hydrogenlikeness is broken (fig 11). However, the relatively small value of photo-current peaks in relation to background, does not enable to observe here the phenomenon as clearly as in CdS.

We established that in the course of time changes take place with the photo-current curves of some CdS crystals; these changes remind one of those that are caused by a mechanical treatment of the surface. First type crystals pass over into 2nd type. A sharp drop of sensitivity in the short-wave part of photo-current curves takes place in 2nd type crystals. This is connected apparently with the deterioration of the surface condition.

An interesting phenomenon is observed at a considerable cooling (to $4^{\circ}K$) of CdS crystals, which at $T = 77^{\circ}K$ belonged to the 2nd type. An increase of sensitivity takes place in the short-wave part of the curves in relation to the long-wave part. Simultaneously the long-wave false peak becomes less clear. However, the photo-current peaks in these crystals

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x/ In our first experiments /15/ when the fact of existence of two types of CdS crystals had not yet been established, we tried to explain the phenomenon of selfreversion of photo-current peaks by the difference of absorption coefficients in ordinary and extraordinary rays. On the ground of the facts set forth, this explanation was rejected as erroneous.

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crystals at T = 4°K correspond to the absorption lines (at T = 4°K) as was the case at T = 77°K.

In some crystals a cooling to 4°K causes a considerable change of the correlation between the photo-current curves and absorption lines. The crystal, which at T = 77°K belonged to the 2nd type (the photo-current dips coincide with the absorption peaks), at T = 4°K passes over into the first type (the photo-current peaks coincide with the absorption lines). The spectral photo-current curves of such a crystal for T = 77°K and T = 4°K are given in fig. 12a.

An account was given above concerning the observed facts relating to the phenomena of photo-conductivity. We find it of some interest to compare them with the phenomena of luminescence, as it is known that both these groups of phenomena are often correlated. It turned out that when studying the spectra of luminescence excitation of CdS crystals (at T = 77°K), a connection between luminescence and exciton absorption lines is also revealed. As is the case on the curves of spectral distribution of photoconductivity excitation, on the curves of luminescence excitations we observe peaks also, which coincide with the exciton absorption lines. This phenomenon has also been observed by Datton /38/, Broude and others /39/.

Recently /41/ it was established that CdS crystals, according to the excitation curves of edge emission (green) luminescence, may be divided, chiefly, into two kinds (fig. 13, curves 4 and 5). In 1st type crystals the peaks on the excitation curves coincide with the exciton absorption lines, and in 2nd type crystals the dips coincide with the exciton absorption lines. These two kinds of crystals are closely connected and

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1st type crystals can be transferred into 2nd type crystals by means of thermal treatment. Thus, an evident parallelism between the phenomena of photoconductivity is observed here.

Similar phenomena in luminescence were discovered in HgI_2 and PbI_2 crystals. In HgI_2 crystals (red modification) the excitation spectra, both of the yellow-green and red luminescence were studied on one and the same sample /40/. The excitation spectra of these luminescence bands considerably differ. If in the case of the excitation spectrum of red luminescence the peaks coincide with the absorption lines ($\lambda\lambda$ 5330A, 4930A), in the excitation spectrum of the yellow-red luminescence, the dips correspond to the absorption lines (fig.13, curves 1, 2 and 3). A surface treatment considerably influences the shape of the exciton curves of yellow-green luminescence; the excitation curves of red luminescence do not noticeably change.

The appearance of photo-current peaks in crystals, where the light in the region of fundimental absorption is absorbed completely by the crystal, is testimony to the existence of two mechanisms of photo-current origination. One of them - the usual one - is connected with the direct formation of free electrons and holes. The second one is connected with light absorption in the lines. The trustworthy fact that the absorption lines appertain to the exciton speaks in favour of the assumption that this second creation mechanism of free carriers has, as an intermediate stage, a neutral particle - the exciton.

Two principal processes of origination of free charges through exciton state are conceived.

1) Dissociation of the exciton by thermal oscillation of the lattice.

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However, the comparatively considerable binding energy of the exciton makes this process hardly probable. Besides, from this point of view, it would be very difficult to explain the phenomenon (which is experimentally observed) of transition of photo-current peaks to dips at thermal treatment of CdS crystals. Furthermore, this process should have had a significant temperature dependence, which, however, is not experimentally observed either in the peaks or the background.x/

2) An interaction of the exciton either with the impurity centre, defect or other excitons. This process provides for a transfer of the exciton energy to the electron of the impurity centre or defect as a result of a "2nd kind collision" /9/. We consider this mechanism of photo-current origination more probable than the first one for the explanation of the phenomena observed by us. The proposed mechanism includes in itself as an obligatory fact the movement of the exciton. Therefore, the existence of peaks on the photo-current curve should then be considered as proof in favour of exciton migration in the crystal lattice.

If we accept this mechanism of origination of photo-current peaks, it must give explanation to the origination of dips on the spectral photoconductivity curve. We assume that the defects in the crystal lattice play here a considerable role. Indeed, the defects are sure to impede the movement of the exciton and stimulate its "premature" nonphotoactive annihila-

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x/ It is possible that in separate cases this mechanism plays a certain role, in the phenomena studied by us, but not a leading one.

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tion. This makes it difficult for the exciton to migrate to those centres, to interact with them and to create free charge carriers. It is natural to assume that the maximum number of such defects will be near the surface. This, we think, may explain the specific shape of the photo-conductivity curves with dips.

The high coefficient in the absorption peak of the exciton lines leads to the origination of excitons chiefly in the thin layer of the crystal in the immediate proximity of the surface. At a large concentration of these defects, the annihilation of excitons will be maximum for spectral regions with the highest absorption coefficient (centre of absorption line). The excitons, formed by the outer parts („wings") of the absorption lines, will be originated in the depth of the crystal, where the concentration of defects is less than near the surface. This must lead to the self-reversion of the exciton photo-current peak on the photo-conductivity curve. This causes the formation of dips and false peaks in 2nd type crystals.

The results of our experiments with thermal treatment and grinding of crystals then receive a natural explanation from the latter point of view. It is known that both at tempering and grinding a large number of defects appear in the crystals. It is not surprising that after these treatments 1st type crystals pass over to 2nd type crystals.

It is natural to assume that annealing "heals" the defective parts of the crystals, and, therefore, the concentration of excitons that take part in the origination of a photo-current, after annealing again increases and photo-current peaks appear again on the places of exciton lines.

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As we have already mentioned earlier, the spectral photo-conductivity curves of the investigated crystals at low temperatures are made up of two parts: photo-current peaks and structureless background. This phenomenon, apparently, is in conformity with the spectrum on the edge of the fundamental absorption, consisting of exciton lines and a continuous background. A rise of the background in photo-conductivity takes place, apparently, simultaneously with the increase of the background in absorption. This is seen, for instance, from our investigations with evaporated CdS films /20/. It must be particularly noted that the intensity of the background on the photo-conductivity curves in relation to the peaks changes not only from sample to sample, but, to a greater degree, from substance to substance. Thus, for instance, in a Cu_2O crystal the height of the peak above the background is so slight that their detection presented certain experimental difficulties. At the same time in CdS crystals the exciton peaksusually stand out boldly. This shows that the contribution of excitons to photo-conductivity may differ greatly from substance to substance.

The photo-electric effectiveness /the quantum yield/ in the lines and background are, probably, different. This is an indication to the different nature of these phenomena. We think that the existence of a background on the photo-conductivity curve is connected with the direct formation of free charge carriers by means of light, i.e., by the ordinary (usually considered) mechanism.

However, one must keep in mind that the primary formation of light of the electron and hole, may lead to the formation of exciton as a secondary phenomenon. We think it is natural

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to assume that a recombination of the electron and hole, may take place through the intermediate formation of exciton, and that, probably, a direct annihilation of the electron and hole is even hardly possible. The electron in a free band and hole when meeting, at first form the exciton in different excitation states, and then annihilate from exciton levels either with emission of light or without radiation. Thus one can assume two ways of exciton formation in a crystal under the effect of light - direct and indirect (through the free electron and hole) /37/. Exciton, formed in such an indirect way, can also cause a photo-current as a result of collision with the impurity centre or phonon. If this phenomenon actually takes place, then the exciton photo-conductivity component may be found not only in peaks but also in a continuous background.

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INSCRIPTIONS TO THE DRAWINGS

Fig.1. Curves of spectral photo-current distribution of two types (1 and 2) of CdS crystals in polarized light.

Fig.2. Curves of spectral photo-current distribution in three CdS crystals at T = 77°K. a - /E⊥C/, b - E//C .

Fig.3. Influence of rotation of a CdS crystal upon the shape of spectral curves of photo-current distribution. a - for 1st type crystals; b - for 2nd type crystals.

Fig.4. The spectral photo-current dependence of a Cu_2O crystal. The tension of applied electric field E = 750 v/cm, T = 77°K (curves a', b',c').

Original curves of a fine structure of spectral photo-current dependence of a Cu_2O crystal. T = 77°K. a - region of first step, E = 1,5 kv/cm.; b - region of exciton yellow series, E = 750 v/cm; c - region of exciton green series, E = 750 v/cm /After Gross and Pasternjak/.

Fig.5. Spectral photo-current dependence of a HgI_2 crystal of red modification at T = 77°K in polarized light.

Fig.6. Spectral photo-current dependence in a HgI_2 crystal of orange modification at T = 77°K.

Fig.7. Spectral photo-current dependence in a PbI_2 crystal at T = 77°K. The crystal optic axis is perpendicular to the sample surface.

Fig.8. Spectral photo-current dependence in a GaSe crystal at T = 77°K. The crystal optic axis is perpendicular to the sample surface.

Fig.9. Spectral photo-current dependence in AgI polycrystalline films. (original curves) a - cubic modification; b - hexagonal modification.

Fig.10. Spectral curves of photo-current distribution (in relative

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units for each curve) of a CdS crystal after different surface treatment at T = 77°K.

a - non-treated crystal (the peaks marked by arrows coincide with absorption lines);

b - after a short polishing on wet silk;

c - after polishing on silk with chromium oxide;

d - after grinding with fine emery-powder.

Fig.11. Spectral photo-conductivity distribution of aCu_2O sample at T = 77°K. Sample thickness - 400μ.

a - natural surface;

b - after etching in HNO_3 (a similar curve, but with a washed out fine structure, is obtained after polishing);

c - after a 20 min. annealing at 150°C in air;

d - after oxygen photo-sorption on the annealed surface.

Shift of the photo-current fine structure peaks in the region of yellow exciton series after surface treatment (original curves).

a' - natural surface;

b' - after etching in HNO_3

/After Gross and Pasternjak /30/.

Fig.12. Spectral photo-current dependence in a CdS crystal at T = 77°K and 4°K in polarized light.

Fig.13. Luminescence excitation spectra of CdS and HgI_2 crystals at T = 77°K. (after Gross, Razbirin and Shekhmametjev / 41 /)

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